IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of Tomoo SUGAWARA
Serial No.: 10/567,257
Filed: February 6, 2006
For: POLYMERIZABLE COMPOSITION AND MOLDED PRODUCT THEREOF

DECLARATION UNDER 37 CFR 1.132

Honorable Commissioner of Patents and Trademarks,
P. O. Box 1450, Alexandria, VA 22313-1450

Sirs:

I, Shigeru FUJITA, a Japanese citizen, residing at 6-2, Marunouni 1-chome, Chiyoda-ku, Tokyo, Japan, hereby declare and state that I have been employed by ZEON Corporation (assignee of the present application) since April 1993. I also declare that I engaged in researching characteristic properties and applications of various specialty rubbers (epichlorhydrin rubbers, hydrogenated acrylonitrile-butadiene rubbers etc.) from July 1993 to October 2005, and I have been engaging in researching ring-opening metathesis polymerization-related technologies since November 2005.

I declare that I have read all of the documents concerning the above-entitled patent application, and am familiar with the contents of the present invention in this application.

I further declare that the following experiments were conducted by myself and that the results of the experiments are all true and correct to the best of my own knowledge.

[Experiments]

Experiments were conducted in accordance with the following items.

1. Object of Experiment
2. Experiments
3. Evaluation

1. Object of Experiment

Further Experiments were conducted to prove that the remarkable effects of the polymerizable composition disclosed in the present inventions could be achieved only by using a monomer having a condensed ring made of an aliphatic ring having one or more carbon-carbon double bonds and an aromatic ring as the cycloolefin monomer and by setting the content of the monomer to 10 % by mass or more in ratio to all the cycloolefin monomers.

2. Experiments

2-1. Experiment 1

(1) Metal Mold

Two chrome plated iron plates in size of 200 mm × 200 mm with a 500 W heater were used. In order to form a gap (cavity) inside between two iron plates, a resin spacer (with a thickness of 4 mm) in the shape of "U" and having the same size as the iron plate was sandwiched between two iron plates, and the 4 corners thereof were clamped with a squill vice. A thermocouple for temperature adjustment was stuck on the cavity-side surface of the upper iron plate in the simple metal mold obtained, and the thermocouple is connected to a temperature adjuster of the heater to control the temperature of the metal mold.

The temperature of the metal mold was set at 70°C.

(2) Polymerizable Composition

Put into a 500 ml polyethylene bottle were 1.1 g of 1,4-dihydro-1,4-methanonaphthalene as a cycloolefin monomer, 98.9g of tetracyclododecene, and 8 g of red phosphorus, 26g of ammonium

polyphosphate and 26g of aluminum hydroxide as a flame retardant. Thereafter, 0.8 ml of toluene solution thereof benzilidene(1,3-dimesitylimiazolidin-2-ylidene)(tricyclohexylphosphine)ruthenium dichloride with a concentration of 0.05 mol/l as a metathesis polymerization catalyst was added into the mixture while stirring to prepare a polymerizable composition.

(3) Molding and Flame Test

The polymerizable composition was poured into the above metal mold at 70°C, and the polymerization reaction progressed exothermically. The molded product was taken out from the metal mold in 3 minutes after pouring the polymerizable composition.

2-2. Experiment 2 (same manner as the Example 1 disclosed in specification)

(1) Metal Mold

Two chrome plated iron plates in size of 200 mm × 200 mm with a 500 W heater were used. In order to form a gap (cavity) inside between two iron plates, a resin spacer (with a thickness of 4 mm) in the shape of "U" and having the same size as the iron plate was sandwiched between two iron plates, and the 4 corners thereof were clamped with a squill vice. A thermocouple for temperature adjustment was stuck on the cavity-side surface of the upper iron plate in the simple metal mold obtained, and the thermocouple is connected to a temperature adjuster of the heater to control the temperature of the metal mold.

The temperature of the metal mold was set at 70°C.

(2) Polymerizable Composition

Put into a 500 ml polyethylene bottle were 60 g of 1,4-dihydro-1,4-methanonaphthalene as a cycloolefin monomer, 140g of

dicyclopentadiene (containing 10% of cyclopentadiene trimer) as other cycloolefin monomer, and 8 g of red phosphorus, 26g of ammonium polyphosphate and 26g of aluminum hydroxide as a flame retardant. Thereafter, 0.8 ml of toluene solution thereof benzilidene(1,3-dimesitylimiazolidin-2-ylidene)(tricyclohexylphosphine)ruthenium dichloride with a concentration of 0.05 mol/l as a metathesis polymerization catalyst was added into the mixture while stirring to prepare a polymerizable composition.

(3) Molding and Flame Test

The polymerizable composition was poured into the above metal mold at 70°C, and the polymerization reaction progressed exothermically. The molded product was taken out from the metal mold in 3 minutes after pouring the polymerizable composition.

2-3. Experiment 3

(1) Put into a 100 ml polyethylene bottle were 45 g of tetracyclododecene, 15g of norbornene and 40 g of phenyl norbornene as a cycloolefin monomer, 12 g of magnesium hydroxide, 4.5 g of melamine polyphosphate and 1.5 g of red phosphorus as a flame retardant, 0.54 ml (0.51 g) of allyl methacrylate as a chain transfer agent, 0.43 ml (0.34 g) of di-t-butyl peroxide (one minute half-life temperature of 186°C) as a radical generating agent, 0.084 g of 3,5-di-t-butylhydroxyanisole as a radical crosslinking retarder and 0.3 g of PLENACT AL-M (aluminate coupling agent, manufactured by Ajinomoto-Fine-Techno Co., Inc.) as a dispersant. Thereafter, 0.31 ml of toluene solution thereof benzilidene(1,3-dimesitylimiazolidin-2-ylidene)(tricyclohexylphosphine)ruthenium dichloride with a concentration of 0.05 mol/l (containing triphenylphosphine at a concentration of 0.25 mol/l) as a metathesis

polymerization catalyst was added into the mixture while stirring to prepare a polymerizable composition.

(2) Two glass cloths (cut off each in size of 200 mm × 200 mm with a thickness of 0.174 mm, with a trade name 7628/AS891AW, manufactured by ASAHI-SHWEBEL CO., LTD) were placed on a glass fiber reinforced PTFE resin film (cut off in size of 300 mm × 300 mm, with a thickness of 0.08 mm, with a product number 5310, manufactured by SAINT-GOBAIN KK), then the polymerizable composition was poured over the glass cloths, then another glass fiber reinforced PTFE resin film same as the above was laminated thereon, and then the laminate was press rolled to be impregnated with the polymerizable composition.

Then, the laminate was placed on a hot plate heated at 145°C for 1 minute to polymerize the monomer. Thereafter, the glass fiber reinforced PTFE resin films adhered to the both surface of the laminate were peeled off to obtain a prepreg.

(3) The three prepregs (cut off each in size of 87 mm × 87 mm) were put into a mold frame in the shape of framed rectangle and in inner size of 90 mm × 90 mm (with a thickness of 1 mm), sandwiched by using PTFE films with a thickness of 0.05 mm, and then the laminate was heat pressed under a pressure of 4.1 MPa at 200°C for 15 minutes. Thereafter, the heat pressed laminate was cooled down while being kept under the pressure and the laminate was taken out from the press after the temperature of cooled down to 100°C or less.

3. Evaluation

3-1. Experiment 1

The UL94 20 mm vertical flame test was conducted to the molded product obtained in Experiment 1, and it was confirmed that the flaming time was 30 seconds or more and thus the molded product was evaluated as

rejectable quality.

3-2. Experiment 2

The UL94 20 mm vertical flame test was conducted to the molded product obtained in Experiment 2, and it was confirmed that the molded product had the evaluation (V-0) of the highest flame retardance.

3-3. Experiment 3

The UL94 20 mm vertical flame test was conducted to the laminate obtained in Experiment 3, and it was confirmed that the flaming time was 30 seconds or more and thus the laminate was evaluated as rejectable quality.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Dated this 8th day of January, 2008

Shigeru Fujita

Shigeru FUJITA